Density of Phonon States at the Fe Sites in Superconducting FeSe as Function of Temperature and Pressure

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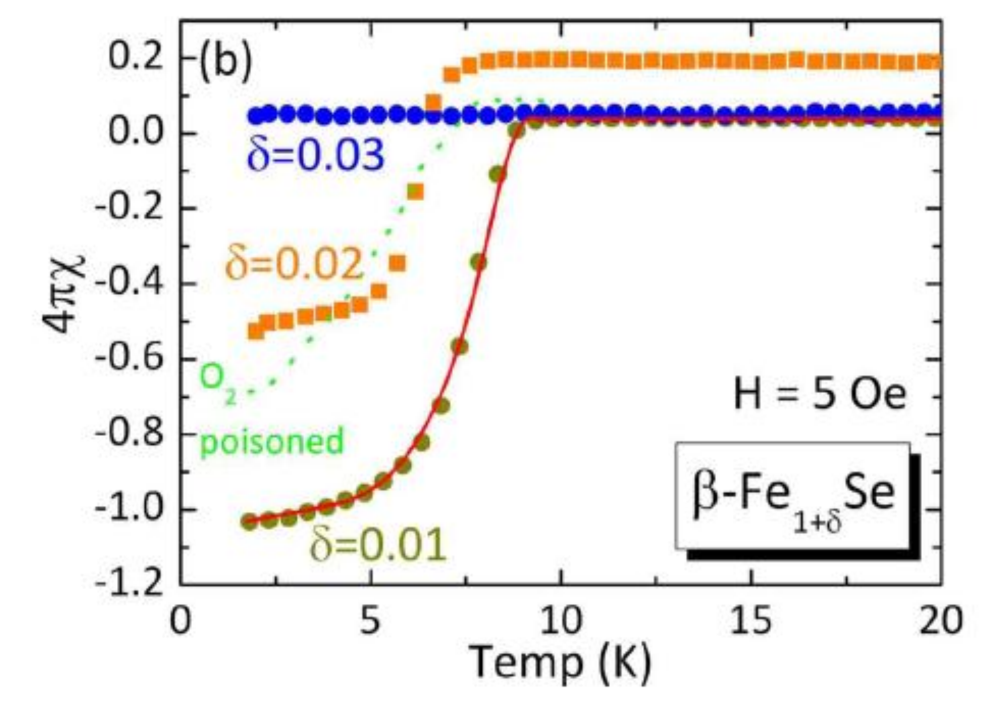
Oral Contribution on the HFI-NQI 2010 at CERN, Geneve, 13.-17.Sept. 2010

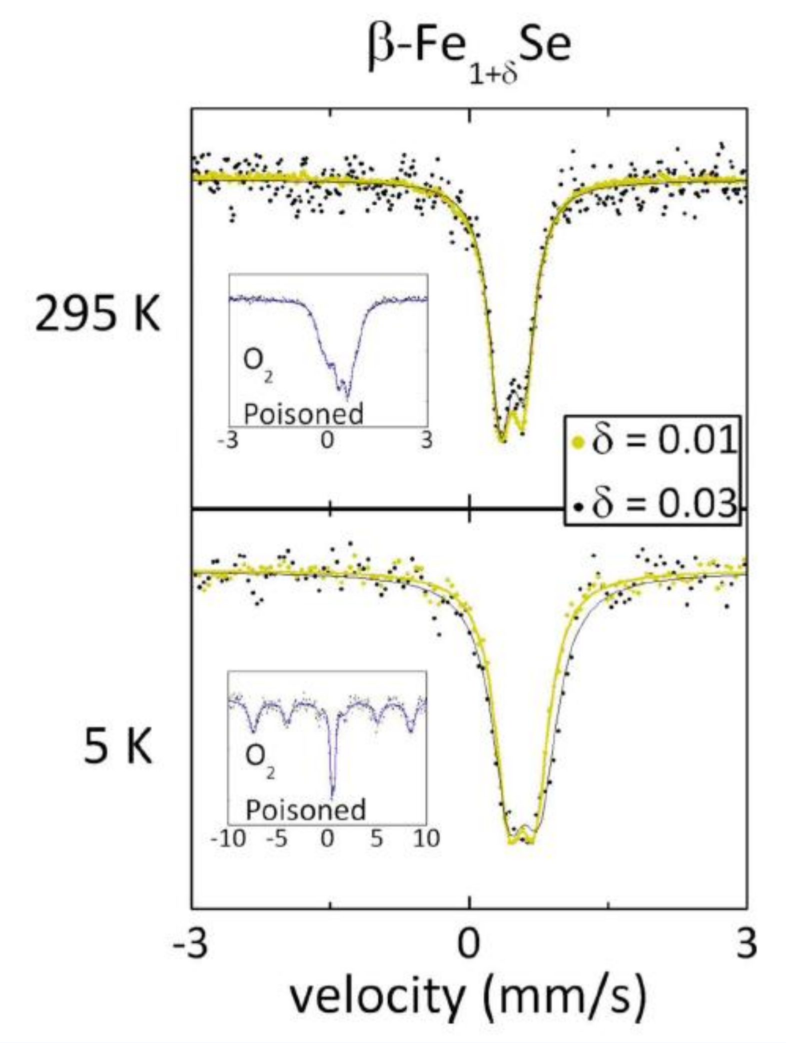
Outline of the talk

- Superconducting (s.c.) $Fe_{1+x}Se$: phase analysis
- Pressure-induced effects in $Fe_{1.01}Se$:
  Superconductivity, structure, Mössbauer spectra
- Temperature and pressure dependence of the density of phonon states at the Fe sites in $Fe_{1.01}Se$
- Comparison with the phonon-DOS in s.c. $FeSe_{0.5}Te_{0.5}$ X-tals: Use of polarisation
- Short discussion of possible pairing mechanism in FeSe systems

Refs: F.C. Hsu, Proc. Natl Acad. Sci. **105**, 14262 (2008).
T.M. McQueen et al., Phys. Rev. B **79**, 014522 (2009).
S. Medvedev et al., Nature Mater. **8**, 630 (2009).

$Fe_{1+\delta}Se$: Characterization



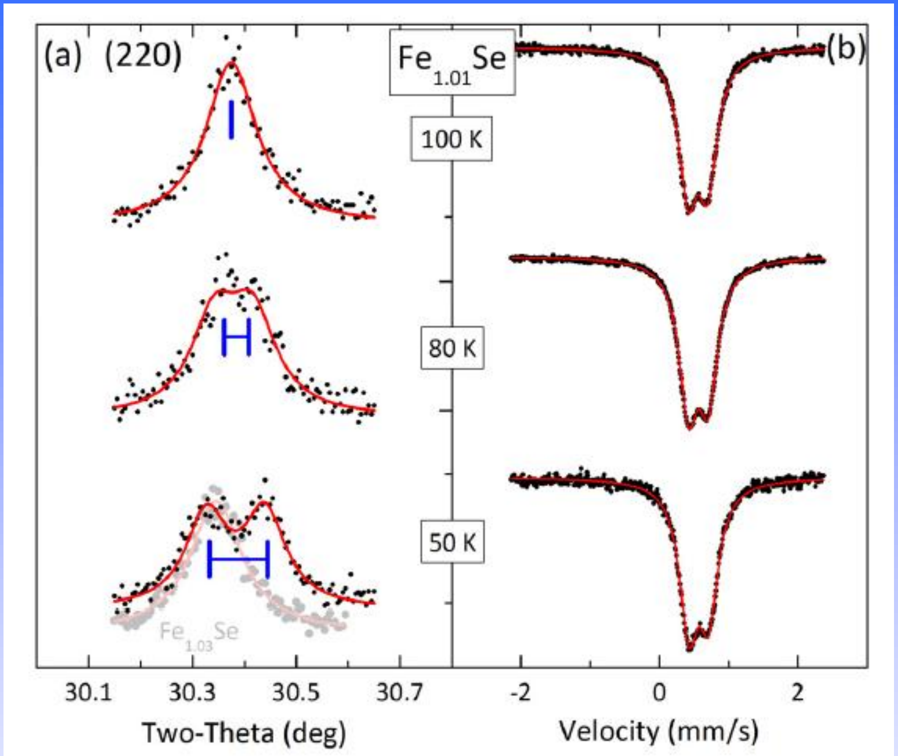


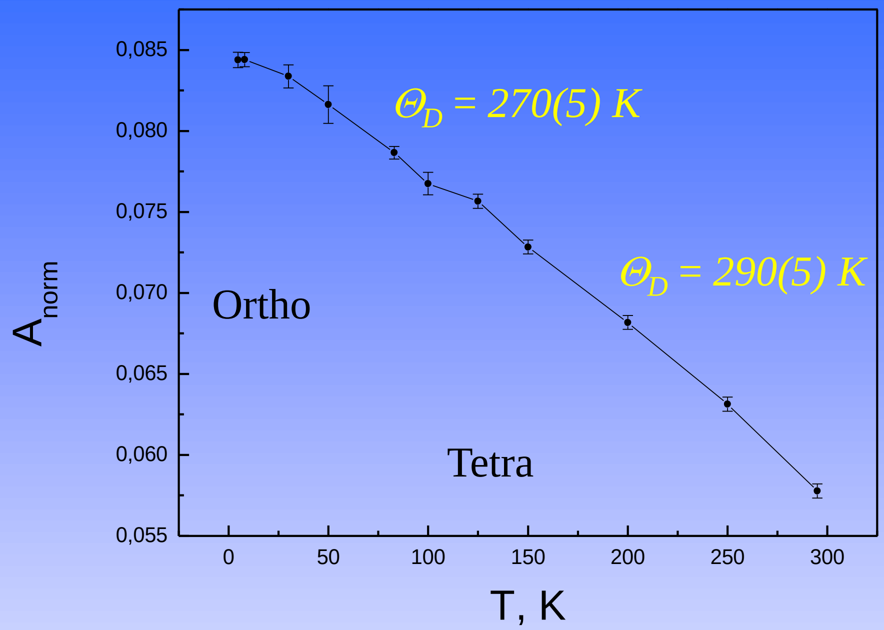
Susceptibiliy and ^{57}Fe-ME studies

T.C. McQueen et al., PRB 79, 014522 (2009).

- Superconductivity (s.c.) in FeSe systems depends sensitively on stoichiometry
- No significant differences in ^{57}Fe-ME spectra between β–$Fe_{1.01}Se$ and β–$Fe_{1.03}Se$
- Both s.c. and non-s.c. samples are non-magnetic, no sign of magnetic ordering

Tetrag.-orthor. transition in $Fe_{1.01}Se$ at ca. 100 K





Normalized area under Mössbauer spectra is sensitive to the tetra.-orthor. transition. Fe binding is slighty softer in orthor. phase. V. Ksenofontov et al., unpublished.

- XRD scans of the (220) reflection in $Fe_{1.01}Se$ show the appearance of the orthorhombic distortion.
- This subtle distortion is not reflected in the ^{57}Fe-Mössbauer quadrupole spectra.

T.M. McQueen et al., PRL **103**, 057002 (2009).

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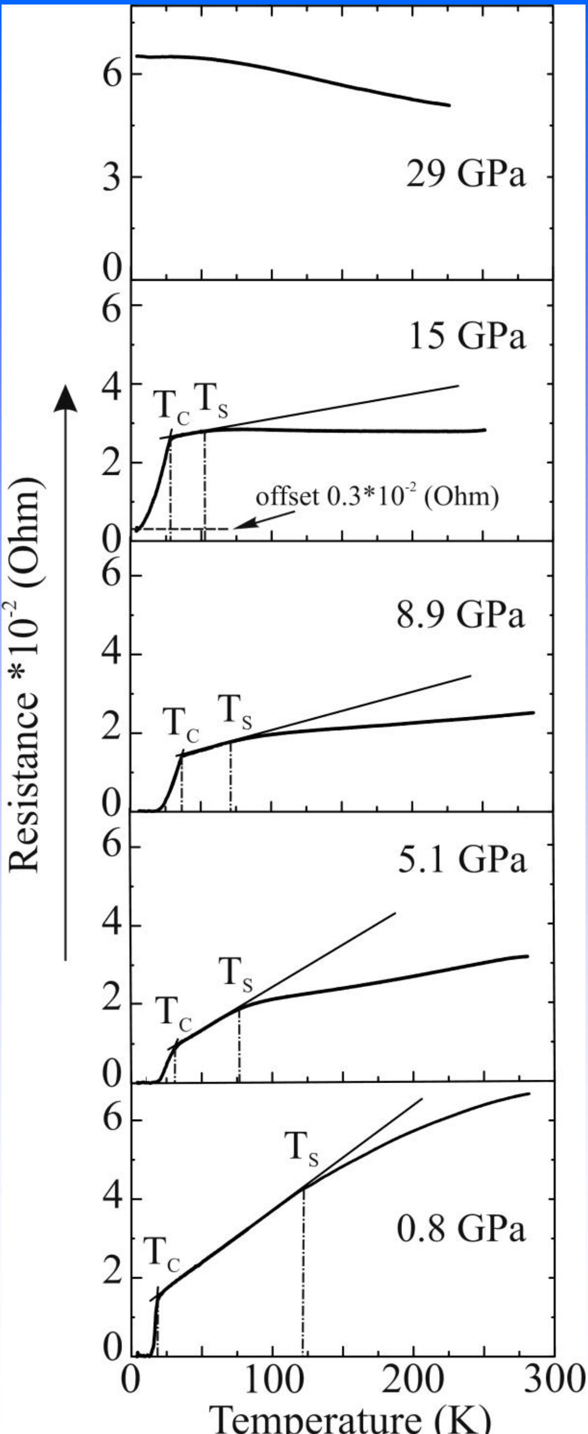
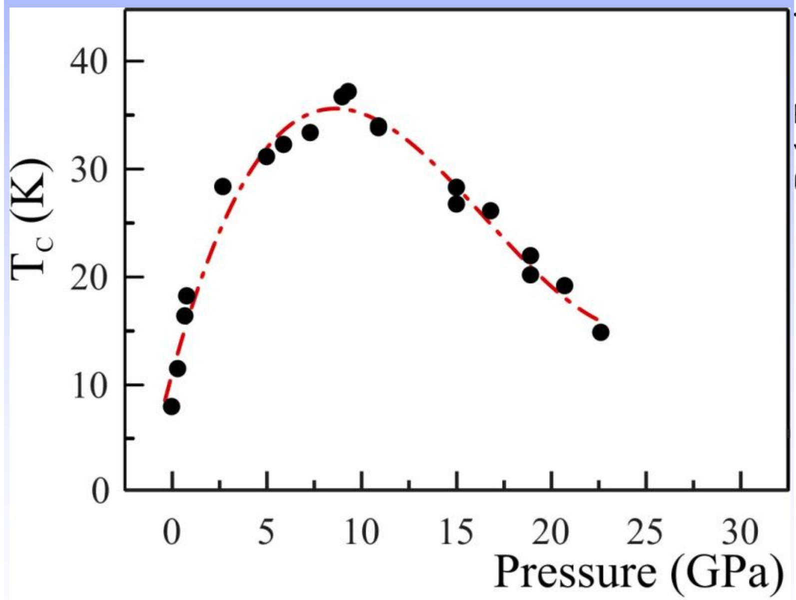


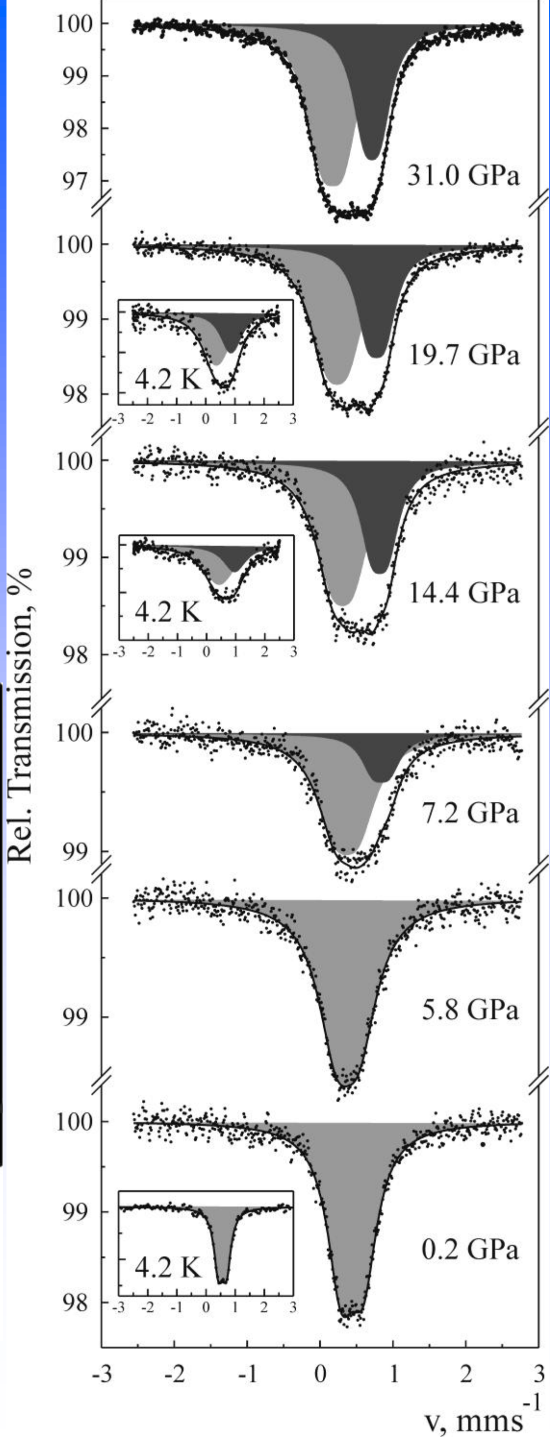
Resistivity and Mössbauer spectra of $Fe_{1.01}Se$ under high pressure

T_c increases from 8.5 K (0 GPa) to 36.7 K at 8.9 GPa

Very steep initial increase: $\delta T_c/\delta p$ = 12.6(8) K/GPa



Ref.: S. Medvedev *et al.*, Nature Mater. 8, 630 (2009)



Short History of Nuclear Resonant Scattering of SR

1958: Detection of the Mössbauer effect
by Rudolf L. Mößbauer

1974: Proposal: Use of SR for MS by
Stan Ruby

1985: Nuclear Bragg Scattering with SR
by Erich Gerdau, Rudolf Rüffer et al.

1991: Nuclear Forward Scattering, NFS (SMS)
J.B. Hastings et al.

meV monochromators

fast detectors (ns)

1995: NFS under pressure (H.F. Grünsteudel et al.)
Magnetism in the Mbar range in RFe_2 (Rainer Lübbers et al.)

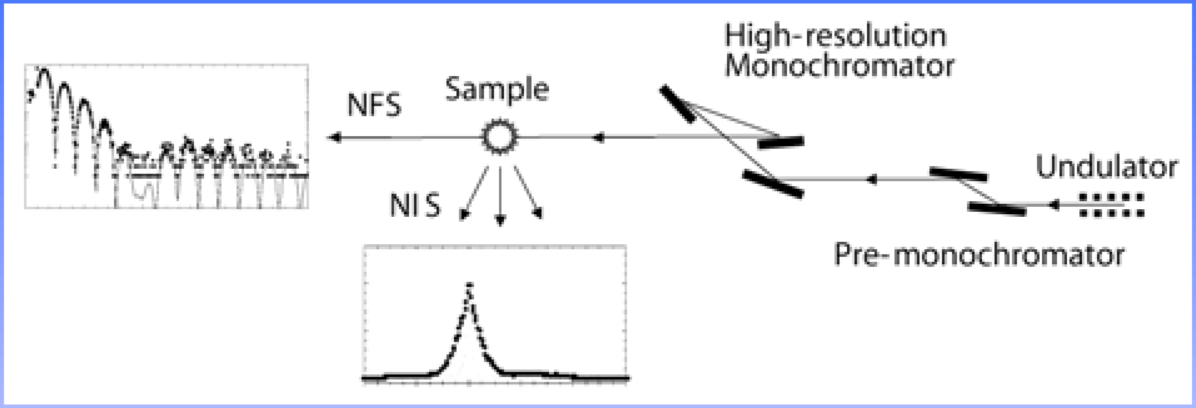
1995: Nuclear Inelastic Scattering, NIS: M. Seto, W. Sturhahn, E.E. Alp
Phonons in iron under pressure with NIS:
2000: R. Lübbers et al., up to 40 GPa, ESRF
2001: H.-K. Mao et al., up to 153 GPa, APS

HP with ^{57}Fe, ^{119}Sn, ^{149}Sm, ^{151}Eu, ..

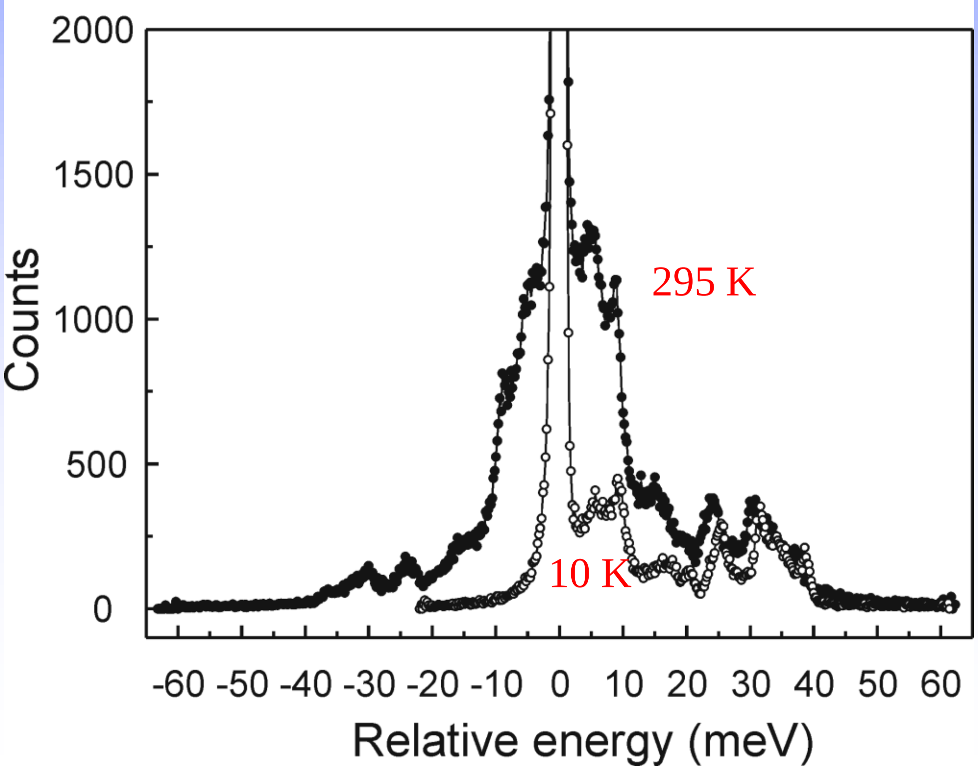
NFS and NIS of synchrotron radiation

NFS: determination of hyperfine interactions

NIS: determination of the local phonon DOS at the Mössbauer atom



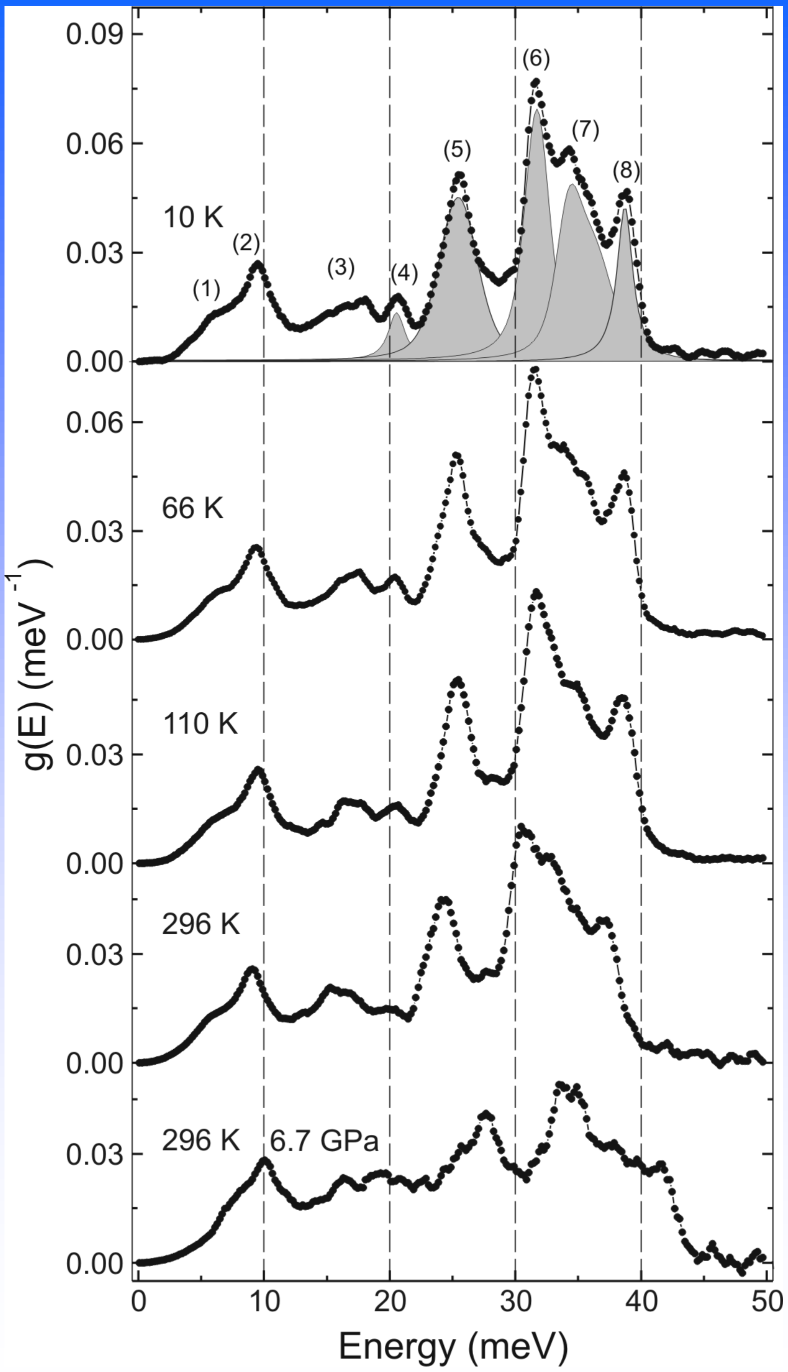
^{57}Fe-NIS studies performed with a resolution of 0.75 meV at 14.4 keV at beamline ID18 of ESRF



^{57}Fe-NIS spectra of $Fe_{1.01}Se$ at 295 K (full circles) and at 10 K (open circles)

V. Ksenofontov, G.W., A.I. Chumakov et al., PRB **81**, 184510 (2010)

^{57}Fe phonon-DOS in sc $Fe_{1.01}Se$ as function of T and P



- **Fe-partial phonon-DOS *g(E)* for $Fe_{1.01}Se$ at different temperatures (above) and at a pressure of 6.7 GPa at 296 K (below). At 10 K, resolved optical modes are indicated, f.i. the B_{1g} Raman mode at 25.5 meV, labeled (5).**

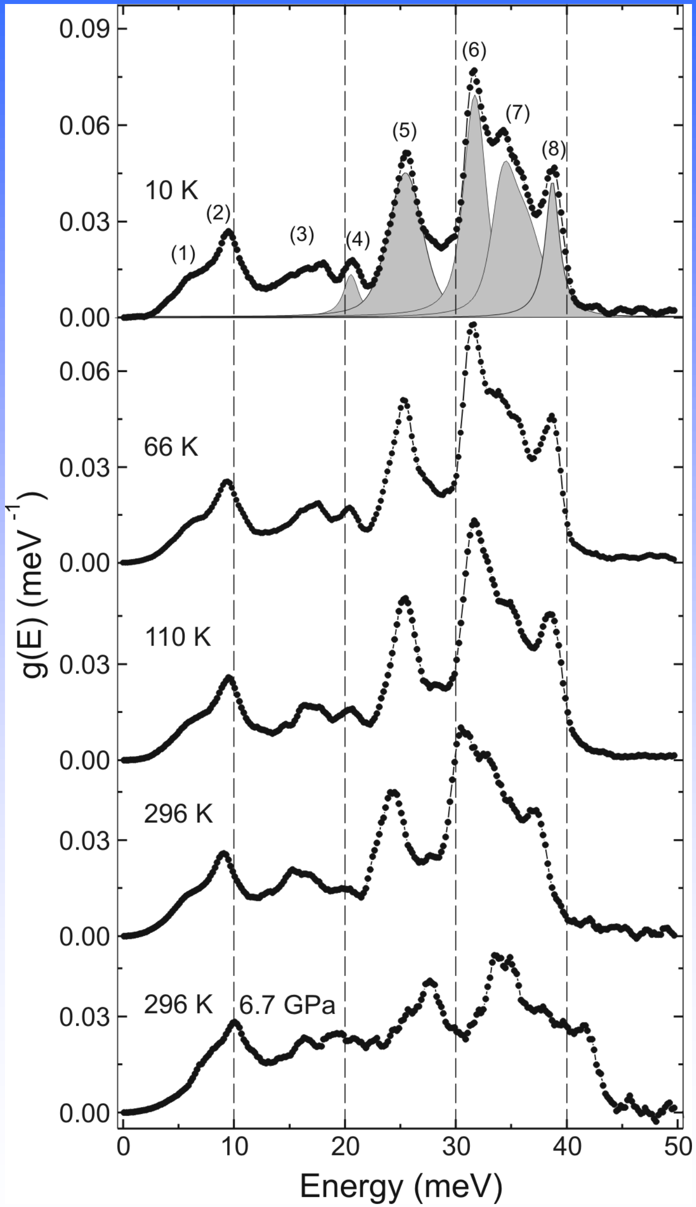
- **Lowering of temperature results in a sharpening of the spectral features and an increase of the mode energies by ~4%.**

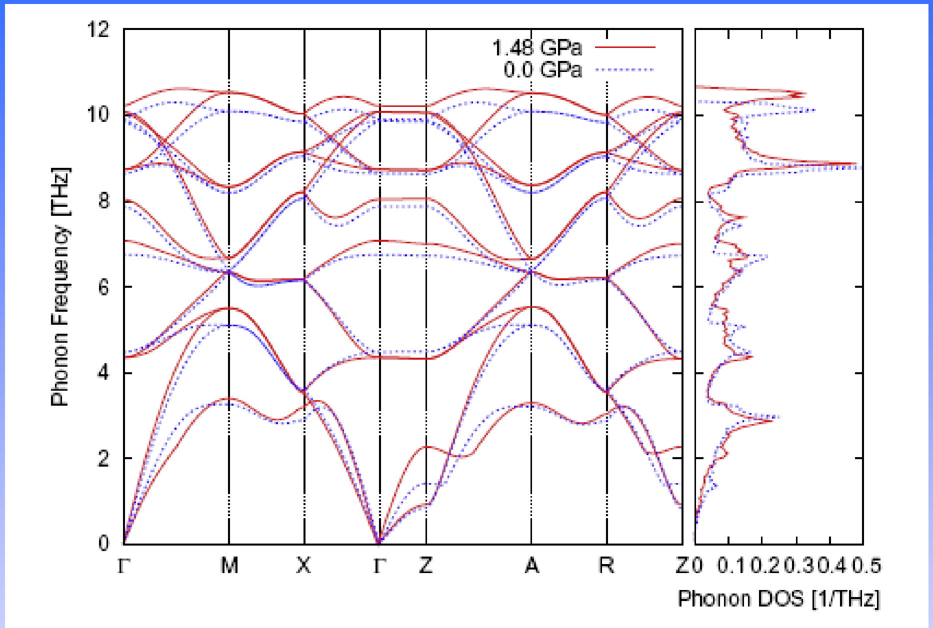
-- No significant changes between the spectra at 110 K (tetr. phase) and 66 K (orthorh. phase).

-- Application of 6.7 GPa (ΔV ~ 14%) at 296 K leads to strong increases of the energies of the optical modes by ~12%, and larger shifts of the acoustic modes by ~30% for mode (1) and ~14% for mode (2).

Ref. 8 of abstract, now published: V. Ksenofontov, G.W., et al., PRB 81, 184510 (2010).

^{57}Fe-NIS studies of local phonon DOS in sc $Fe_{1.01}Se$: Comparison with theory and INS





- Above: Calculated dispersion relations and total phonon-DOS in FeSe by N. Nakamura et al., Physica C 469, 1024 (2009).
- Similar calculations: A. Subedi et al., PRB 78, 134514 (2008).
- The optical modes are quite well described, energies ~10% too large, acoustic mode energies differ considerably due to soft c-axis with complex interlayer interactions, similar to ^{119}Sn-NIS studies of SnO with the same PbO-structure: H. Giefers et al., PRB 74, 094303 (2006).
- Present ^{57}Fe-NIS data compare well with an INS study of FeSe, measuring the *total* phonon-DOS as function of temperature: D. Phelan et al., PRB 78, 134514 (2008).

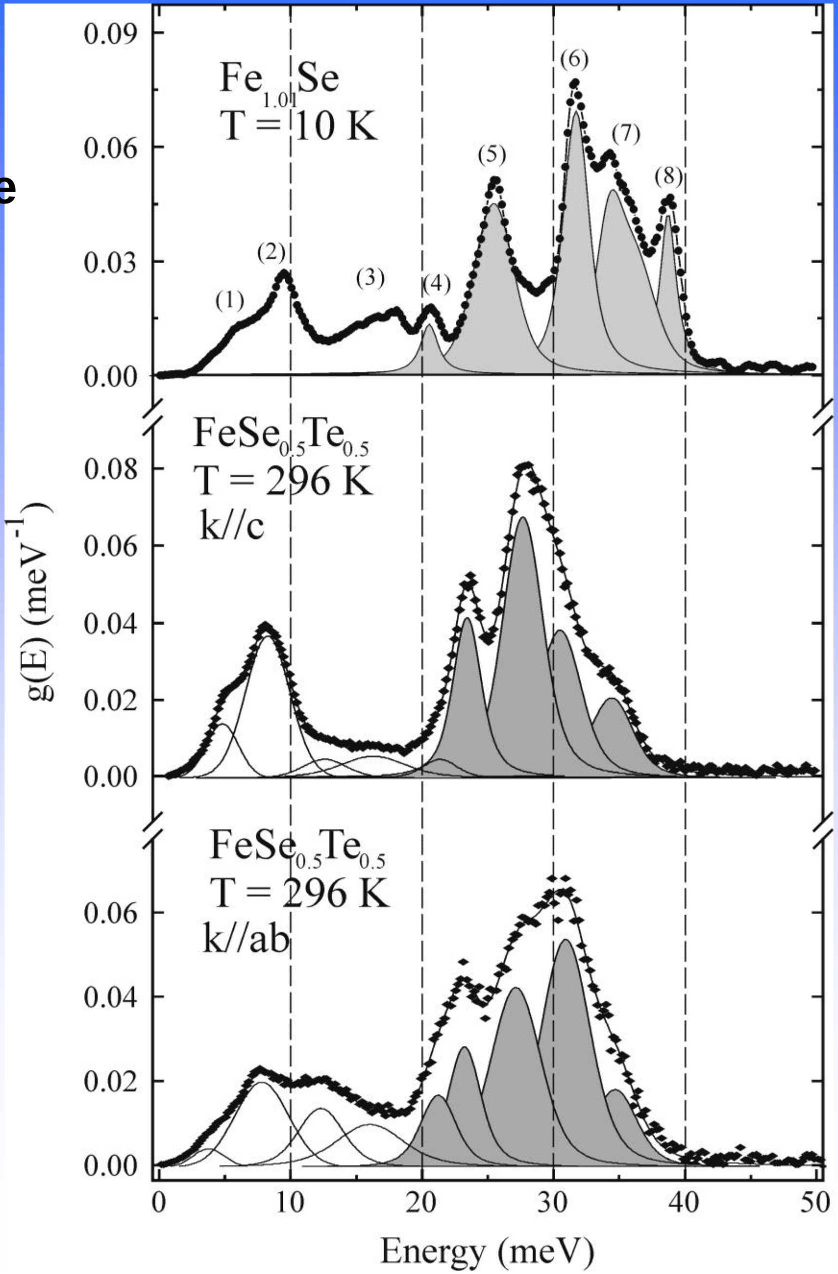
Fe phonon-DOS in FeSe compared with $FeSe_{0.5}Te_{0.5}$ X-tals*

*100% enriched in ^{57}Fe, from V. Tsurkan, J. Deisenhofer, A. Loidl (Univ. Augsburg)

FeSe at 10 K, polycryst. sample with texture
T_C = 8 K

$FeSe_{0.5}Te_{0.5}$ X-tal
k // c, 296 K
T_C = 14 K

$FeSe_{0.5}Te_{0.5}$ X-tal
k // ab, 296 K
T_C = 14 K



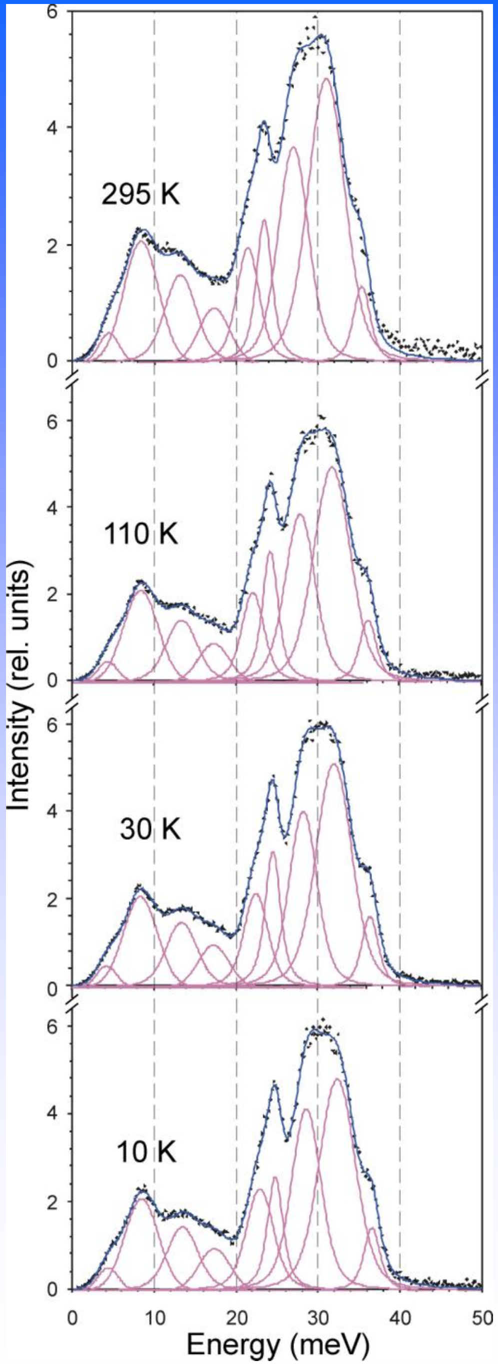
Optical modes adjusted by 5 Gaussians (#4-9): PRB 81, 184510 (2010).

By Te substitution, strong shift of all modes to lower energy, caused by combined (not additive) effcts of:
- lattice expansion of ~10% (negative pressure of ~5 GPa)
- increase of the effective mass m* by ~8%.

All spectral features of FeSe present, but with strong polari-sation dependence, allowing an assignment of modes. Spectral features are broadened by Te substitution and by higher temperature (296 K).

V. Ksenofontov et al., unpubl.

Fe phonon-DOS in $FeSe_{0.5}Te_{0.5}$: temperature dependence



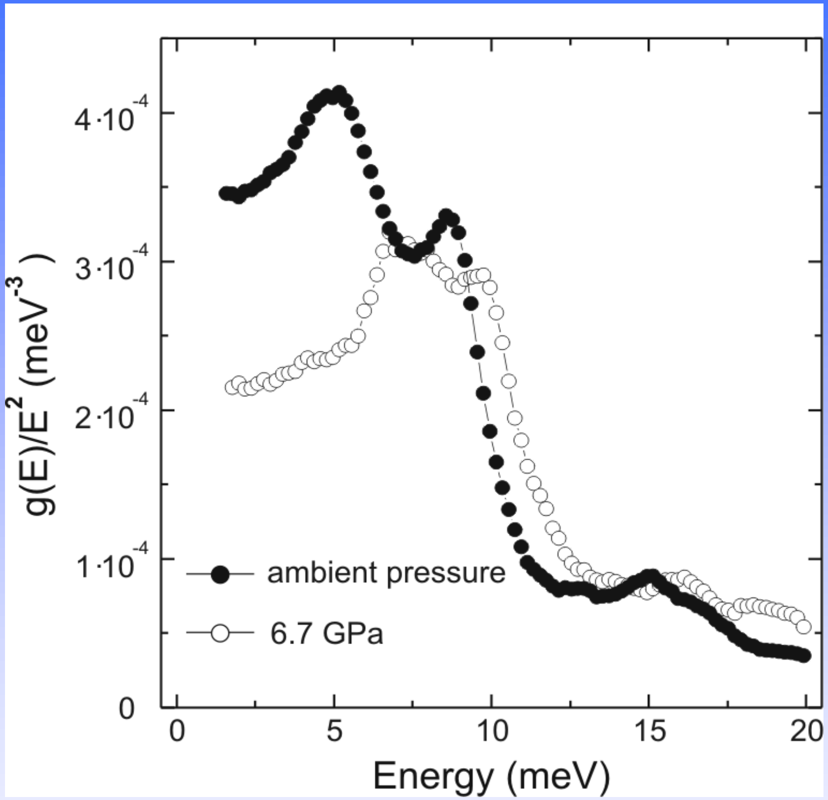
$FeSe_{0.5}Te_{0.5}$ polycryst sample with strong texture, as evident from X-tal sample with $\gamma \perp$ c-axis.

Same principal behaviour as for T-dependent phonon-DOS of FeSe:
(i) sharpening of spectral features and
(ii) shift to higher energies by ~4% by lowering temperature from 295 K to 10 K.

Subtle changes in phonon-DOS above/below T_c = 14 K and above below tetr./orthorh. p.t. are still under investigation.

V. Ksenofontov et al., in preparation.

^{57}Fe reduced phonon-DOS, $g(E)/E^2$ vs. E, in sc $Fe_{1.01}Se$ as function of pressure, and derived elastic properties



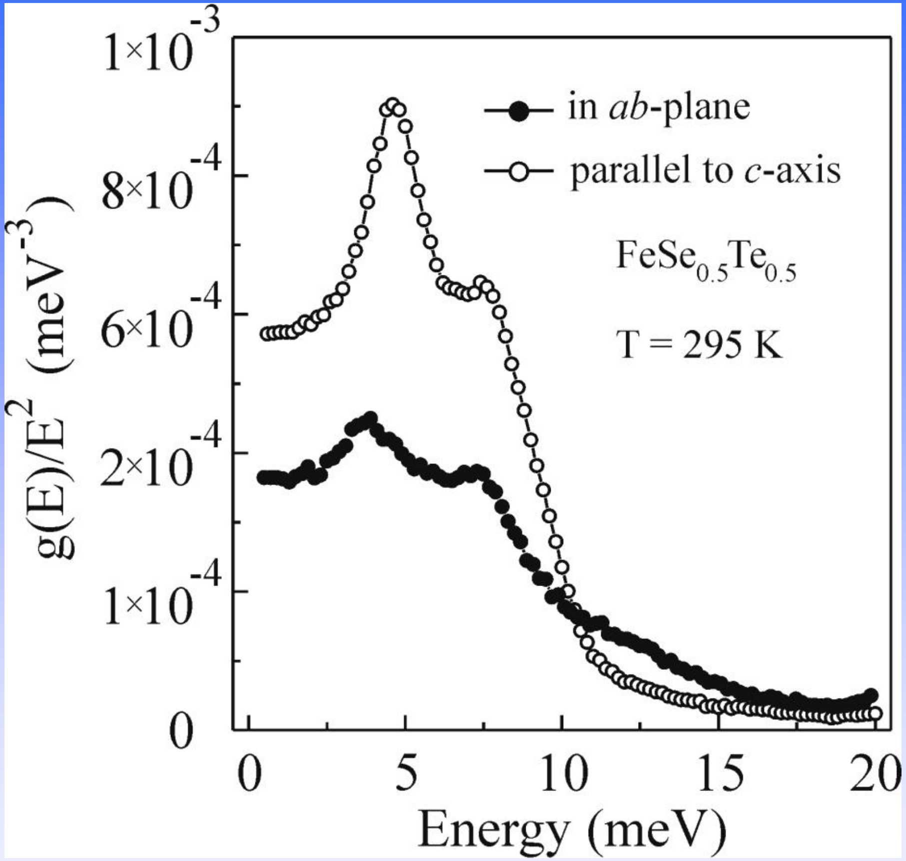
$$\lim_{E\to 0}\frac{g(E)}{E^2}=\alpha=\left(\frac{2m}{m+M}\right)\frac{1}{2\pi^2\hbar^3 n\langle v_D\rangle^3}$$

$$\Theta_{D,LT}=(3/a)^{1/3}/k_B$$

ambient pressure	6.7 GPa
$\Theta_{D,LT}$ = 240(5) K	$\Theta_{D,LT}$ = 278(8) K
$\langle v_D\rangle$ = 2.05(4) km/s	$\langle v_D\rangle$ = 2.25(7) km/s
f_{LM} = 0.60(1)	f_{LM} = 0.66(1)
Θ_D = 285(4) K	Θ_D = 317(6) K

Fe-partial phonon-DOS, $g(E)/E^2$ for $Fe_{1.01}Se$ at 295 K, demonstrating for the low-energy range the strong modification of the lowest acoustic mode at 5 meV under pressure, attributed to vibrations along the “soft” c-axis.

^{57}Fe reduced phonon-DOS, $g(E)/E^2$ vs. E, in sc $FeSe_{0.5}Te_{0.5}$ as function of crystal orientation



γ // c-axis	γ ⊥ c-axis
$\Theta_{D,LT}$ = *200(5) K*	$\Theta_{D,LT}$ = 233(5) K
$\langle v_D \rangle$=1.67(6)km/s	$\langle v_D \rangle$= 1.94(5) km/s
f_{LM} = 0.457(6)	f_{LM} = 0.576(5)
Θ_D = 229(7) K	Θ_D = 273(5) K

- Fe-partial phonon-DOS, $g(E)/E^2$, for $FeSe_{0.5}Te_{0.5}$ at 295 K, demonstrating the strong modification of the acoustic modes in different X-tal directions:
- The “soft” mode, now at 4.5 meV, reflects the high c-axis compressibility.
- Preliminary evaluation, shown is the projected phonon-DOS with g(E) ~$\cos^2$(k,c)

Pressure effect on T_c in $Fe_{1.01}Se$

$$T_C = \frac{\Theta_D}{1.45} \exp\left[-\frac{1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right]$$

$\mu^* \cong 0.1$ - Coulomb pseudopotential

Θ_D = 285(4) K

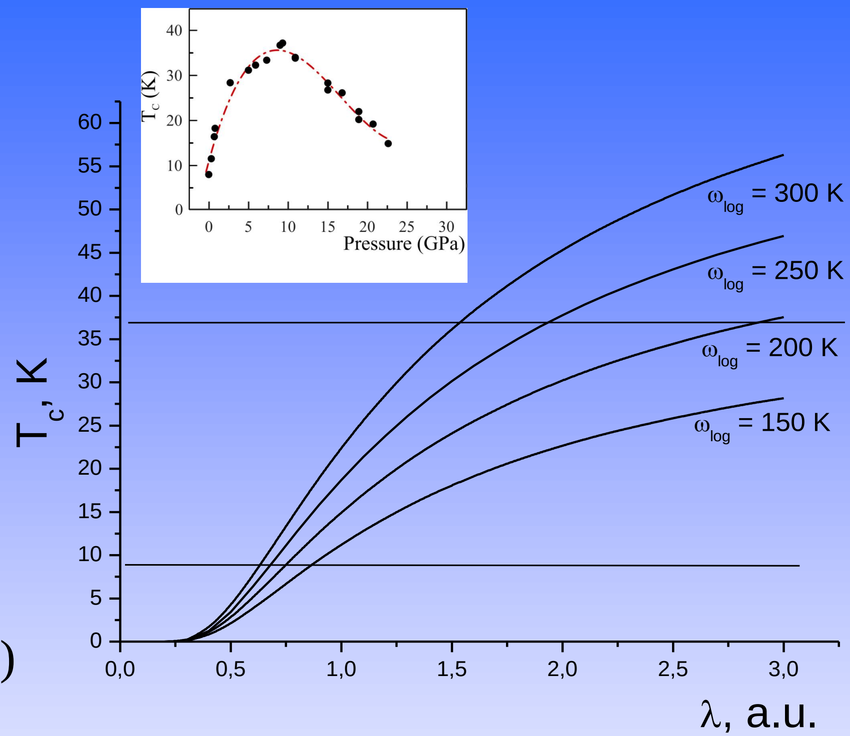
$T_c \cong$ 8.0 K corresponds to λ = 0.65(1)

Electron-phonon coupling constant $\lambda \sim \langle\omega^{-2}\rangle$

Theory: λ = 0.1 (D. J. Singh)

Experiment: λ = 0.21 for LaOFeAs (L. Boeri)

λ = 1.3 for $PrFeAsO_{1-x}F_x$ (D. Bhoi)



Pronounced increase of T_c in $Fe_{1.01}Se$ with pressure cannot be described in the framework of classical electron-phonon coupling, which accounts for only ~15%.

⇒ *Unconventional superconductivity* connected with:

- afm spin fluctuations (^{77}Se-NMR): T. Imai et al., PRL 102, 177055 (2009).

- Se (As) anion height: H. Okabe et al., arXiv:1002.1832v2 (2010).

- Fe-Se bonding angle

A. Subedi, L. Zhang, D.J. Singh, M.H. Du, Phys. Rev. B **78**, 134514 (2008).

L. Boeri, O.V. Dolgov, and A.A. Golubov, Phys. Rev. Lett. **101**, 026403 (2008).

D. Bhoi, P. Mandal, and P. Choudhury, Supercond. Sci. Technol. **21**, 125021 (2008).

Conclusions

- ^{57}Fe-NIS spectra of s.c. FeSe and $FeSe_{0.5}Te_{0.5}$ deliver detailed information about the temperature and pressure dependence of the local phonon-DOS, which can be used to prove theoretical calculations of these properties.

- Pressure effects on phonon-DOS of $Fe_{1.01}Se$ alone does not explain the extremely strong increase of T_c with pressure: Unconventional superconducvtivity.

-Single X-tals of $FeSe_{0.5}Te_{0.5}$ demonstrate anisotropic elastic properties of the lattice and can be used for an assignment of different modes.

-Future work, also on FeSe X-tals, will be devoted to unravel possible connection of certain phonon modes with s.c. mechanisms.

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Department Chemistry, Princeton:

Tyrel M. McQueen, R.J. Cava ($Fe_{1.01}Se$ samples)

Institut Physik, Uni Augsburg:

V. Tsurkan, J. Deisenhofer. A. Loidl ($FeSe_{0.5}Te_{0.5}$ samples)

Thank you for your attention